

# Uncertainty and spatial variability in characterization factors for aquatic acidification at the global scale

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## Abstract

**Purpose** Characterization factors (CFs) quantifying the potential impact of acidifying emissions on inland aquatic environments in life cycle assessment are typically available on a generic level. The lack of spatial differentiation may weaken the relevance of generic CFs since it was shown that regional impact categories such as aquatic acidification were influenced by the surroundings of the emission location. This paper presents a novel approach for the development of spatially differentiated CFs at a global scale for the aquatic acidification impact category.

**Methods** CFs were defined as the change in relative decrease of lake fish species richness due to a change in acidifying chemicals emissions. The characterization model includes the modelling steps linking emission to atmospheric acid deposition (atmospheric fate factor) change, which lead to lake  $H^+$  concentration (receiving environment fate factor) change and a decrease in relative fish species richness (effect factor). We also evaluated the significance of each factor (i.e. atmospheric fate, receiving environment fate and effects) to the overall CFs spatial variability and parameter uncertainty.

**Results and discussion** The highest CFs were found for emissions occurring in Canada, Scandinavia and the northern

central Asia because of the extensive lake areas in these regions (lake areas being one of the parameters of the CFs; the bigger the lake areas, the higher the CFs). The CFs' spatial variability ranged over 5, 6 and 8 orders of magnitude for  $NO_x$ ,  $SO_2$  and  $NH_3$  emissions, respectively. We found that the aquatic receiving environment fate factor is the dominant contributor to the overall spatial variability of the CFs, while the effect factors contributed to 98 % of the total parameter uncertainty.

**Conclusions** The resulting characterization model and factors enable a consistent evaluation of spatially explicit acidifying emissions impacts at the global scale.

**Keywords** Acidification · Atmospheric fate · Characterization factors · Ecological effects · Endpoint · Soil sensitivity · Spatial variability · Uncertainty analysis

## 1 Introduction

Inland surface water acidification has been recognized as a major environmental problem in many parts of Europe and North America for several decades (Skjelkvale et al. 2005). Surface water acidification is the result of the atmospheric emissions of acidifying substances [i.e. nitrogen oxides ( $NO_x$ , i.e.  $NO$ ,  $NO_2$ ,  $NO_3$  and  $HNO_2$ ), ammonia ( $NH_3$ ) and sulphur dioxide ( $SO_2$ )] and their subsequent direct or indirect deposition on aquatic environment. The latter stems from deposition on terrestrial environments that travels to aquatic environments through runoff or underground water (Jeffries and Ouimet 2004). Typically, acidity alone is not toxic to aquatic ecosystem species (e.g. fish, aquatic plants and aquatic insects), but it creates a number of indirect consequences such as a decrease in nutrients or an increase in soluble toxic metallic substances (e.g. aluminum). Hence, for nearly every living organism (whether they are terrestrial or aquatic), a pH optimum exists, and deviation from this

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optimum affects the species richness of the aquatic ecosystem (Jeffries and Ouimet 2004).

Life cycle assessment (LCA) typically evaluates the potential impacts of acidifying emissions on inland surface water through generic characterization factors (CFs) (Heijungs et al. 1992; Humbert et al. 2004; Guinée et al. 2001). The lack of spatial differentiation may weaken the relevance of generic CFs since it was shown that regional impact categories such as aquatic acidification were influenced by the surroundings of the emission location (Potting and Hauschild 2006). Due to this lack of relevance, the generic factors were not integrated to regionalized impact assessment methods (Hauschild et al. 2011). Outside of LCA, the potential impacts of lake acidification are typically evaluated with critical loads (Posch 2004), which are defined as “quantitative estimates of exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Kuylenstierna et al. 2001). Typical critical load calculations are based on surface water modelling, which relies on current surface water input data. An alternative is to use soil modelling that captures key long-term interactions in aquatic environments (Rapp and Bishop 2003; Downing et al. 1993). In LCA, however, the question as to whether or not it is appropriate to use critical load approaches is still open to debate (Hauschild et al. 2011).

It has been recognized that uncertainty evaluation is required to support LCA result interpretation, but, so far, uncertainty evaluation is still not considered in aquatic acidification characterization models or characterization factors (Finnveden et al. 2009).

This paper aims to (a) develop a characterization model and spatially explicit CFs to evaluate potential global-scale impacts on aquatic environments by combining three assessment steps: atmospheric fate, receiving environment fate and ecological effects and (b) evaluate the contribution of these assessment steps to overall CFs spatial variability and parameter uncertainty.

## 2 Methods

### 2.1 Characterization factors

CFs for aquatic acidification (Eq. 1) assesses the potential impacts of acidifying emissions to lakes. CFs are defined as the change in potentially disappeared fraction of fish species (PDF) in lake(s)  $k$  due to a change in atmospheric deposition in the receiving environments in grid  $g$  (i.e. both terrestrial and aquatic environments) following a marginal change in the atmospheric emissions of acidifying substance  $p$  at source location  $i$ . The endpoint CF may be subdivided into an

atmospheric fate factor (FF), a receiving environment fate factor (RF) and an effect factor (EF). CFs are expressed in  $[\text{PDF} \times \text{m}^2 \times \text{year} \times \text{kg}_{\text{emitted}}^{-1}]$  and were calculated for 13, 104  $2^\circ \times 2.5^\circ$  spatial resolution emissions grids.

$$\text{CF}_{i,p} = \sum_g \left( \text{FF}_{i,g,p} \times \sum_g (\text{RF}_{g,k,p} \times \text{EF}_k) \right) \quad (1)$$

### 2.2 Atmospheric fate factors

The atmospheric fate factor (Eq. 2;  $[\text{keq} \times \text{kg}_{\text{emitted}}^{-1}]$ ) describes the link between the change in pollutant emission in grid cell  $i$  of acidifying pollutant  $p$  to the change in acid deposition on grid  $g$ . Atmospheric fate factors were taken from Roy et al. (2012b) who calculated worldwide source receptor matrices whose elements are spatially explicit  $2^\circ \times 2.5^\circ$  atmospheric fate factors.

$$\text{FF}_{i,g,p} = \frac{dD_{g,p}}{dE_{i,p}} \quad (2)$$

Where  $D_{g,p}$   $[\text{keq} \times \text{year}^{-1}]$  is pollutant  $p$  deposition in receiving environment  $g$  related to emission  $E_{i,p}$   $[\text{kg NO}_x, \text{NH}_3 \text{ or } \text{SO}_{2\text{emitted}} \times \text{year}^{-1}]$  at location (air grid cell)  $i$ .

Roy et al. (2012b) based their calculations on the yearly averaged results from a GEOS-Chem  $2^\circ \times 2.5^\circ$  grid spatial resolution simulation for reference year 2005, which is representative of the meteorological average from 1961 to 1990 according to the National Oceanic and Atmospheric Administration's National Climatic Data Center (NOAA National Climatic Data Center 2005).

GEOS-Chem is a global 3D model of tropospheric chemistry driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Data Assimilation Office (Bey et al. 2001). GEOS-Chem simultaneously considers all emissions, transport, chemical transformations and deposition processes on a global level. The implemented wet deposition scheme (Jacob et al. 2000) considers the direct deposition from scavenging in wet convective updraft, the in-cloud scavenging of the pollutant by precipitation of cloud droplets or ice crystals and the below cloud scavenging by precipitation with possible re-evaporation. The default dry deposition was also used (Bey et al. 2001). All calculated deposition (wet + dry) values refer to those reaching ground level. Various kinetic and photochemical reactions are also considered in GEOS-Chem (Evans and Jingqiu 2009). All relevant input data are provided with GEOS-Chem.

### 2.3 Receiving environment fate factors

The receiving environment fate factors (RF (Eq. 3);  $[\text{mol L}^{-1} \times \text{m}^2 \times \text{keq}^{-1} \text{ year}]$ ) evaluate the change in  $\text{H}^+$

concentration in lake(s)  $k$  according to a change in deposition of pollutant  $p$  at location  $g$ . RFs were calculated for every receiving environment within grids  $g$  at a worldwide spatial resolution of  $0.5^\circ \times 0.5^\circ$ . We modelled changes in pollutant deposition that follow a relative 10 % increase in emissions, which is commonly used in LCIA studies (Huijbregts et al. 2000; Krewitt et al. 2001; Potting et al. 1998).

$$RF_{i,g,p} = \frac{\sum_k dC_{g,k} \times A_k}{dD_{g,p}} \approx \frac{\sum_k \Delta C_{g,k} \times A_k}{\Delta D_{g,p}} \quad (3)$$

Where  $C_{g,k}$  [mol  $H^+ \times L^{-1}$ ] is lake  $k$   $H^+$  surface concentration due to deposition at location  $g$  ( $D_{g,p}$ ) and  $A_k$  the lake  $k$  area

$$\begin{aligned} & \underbrace{(C_{a,g} \times P_{a,g} \times LA_g) + (C_{t,g} \times R_{t,g} \times TA_g) + (C_{g-1} \times Q_{g-1})}_{\text{IN}} + \underbrace{r_{g,k} \times V_{g,k}}_{\text{PRODUCTION}} \\ &= \underbrace{C_{g,k}^{\text{mean}} \times Q_{g+1}}_{\text{OUT}} - \underbrace{\frac{dC_{g,k}^{\text{mean}}}{dt}}_{\text{ACCUMULATION}} \end{aligned} \quad (4)$$

Where  $C_{g,k}^{\text{mean}}$  [mol  $H^+ L^{-1}$ ] is the  $H^+$  mean water concentration in grid  $g$ .  $C_{a,g}$  [mol  $H^+ L^{-1}$ ] is the atmospheric deposits  $H^+$  concentration,  $P_{a,g}$  [dm<sup>2</sup>/year] is the precipitation levels,  $LA_g$  [dm<sup>2</sup>] the lakes total area in grid  $g$ ,  $C_{t,g}$  [mol  $H^+ L^{-1}$ ] the grid  $g$  soil solution  $H^+$  concentration travelling to surface waters,  $R_{t,g}$  [dm/year] the runoff from soils in grid  $g$ ,  $TA_g$  [dm<sup>2</sup>] the terrestrial area of grid  $g$ ,  $r_{g,k}$  the grid  $g$  lakes  $k$  production/neutralising term,  $V_{g,k}$  the volume of lakes  $k$  in grid  $g$ ,  $C_{g-1}$  [mol  $H^+ L^{-1}$ ] the upstream  $H^+$  water concentration,  $Q_{g-1}$  [L/year] the input (upstream) flow and  $Q_{g+1}$  [L/year] the output (downstream) flow. Input and output directions of a specific  $0.5^\circ \times 0.5^\circ$  grid were identified in the Vörösmarty et al. (2000a, 2000b) river network.

We then omitted the biogeochemical reactions expressed by the production/neutralising term ( $r_{g,k} \times V_{g,k}$ ) due to global scale data limitations and the fact that, at least in the eastern part of Canada, these reactions are insignificant over a long period of time in comparison to soil watershed inputs (Jeffries and Ouimet 2004). The repercussions of neglecting this production/neutralising term on the model results are detailed in the discussion.  $C_{g,k}^{\text{mean}}$  is obtained by solving Eq. 5.

$$C_{g,k}^{\text{mean}} = \left[ \frac{(C_{a,g} \times P_{a,g} \times LA_g) + (C_{t,g} \times R_{t,g} \times TA_g) + (C_{g-1} \times Q_{g-1})}{(Q_{g+1})} \right] \quad (5)$$

[m<sup>2</sup>]. The lake areas were obtained from the Global Lakes and Wetlands database levels 1 (lake area  $\geq 50$  km<sup>2</sup>) and 2 ( $50$  km<sup>2</sup> > lake area  $\geq 0.1$  km<sup>2</sup>) (Lehner and Döll 2004). Since the lake areas were geo-referenced at their centres and could encompass many  $0.5^\circ \times 0.5^\circ$  grids, we allocated each lake area to their respective  $0.5^\circ \times 0.5^\circ$  grids using geographic information system (GIS) software (ArcMap 9.3).

To calculate  $C_{g,k}$ , we modelled each grid  $g$  (spatial resolution  $0.5^\circ \times 0.5^\circ$ ) as continuously stirred tank reactor (CSTR) operating under steady state conditions ( $dC/dt=0$ ). This implies a homogeneous  $H^+$  concentration ( $C_{g,k}^{\text{mean}}$ ) within the grid lakes. We performed a mass balance for each grid  $g$ . Equation 4 presents the general mass balance equation.

$C_{g,k}^{\text{mean}}$  must be corrected to express the lake surface concentration and be coherent with the effect factor species richness regressions. Indeed, these regressions rely on surface  $H^+$  concentrations, not mean water concentration. Therefore, we empirically corrected  $C_{g,k}^{\text{mean}}$  by assuming a worldwide average lake depth of 10 m [such as considered by Kourzeneva (2009)] and a difference of three on the pH scale between the surface (0–1 m) and the bottom ( $\approx 10$  m) of the lake based on observations (at two different lakes) by Araoye (2009) and by Wang and Chen (1990). According to these assumptions,  $C_{g,k}^{\text{mean}}$  would then represent the pH at a depth of 5 m, and the lake surface  $H^+$  concentration ( $C_{g,k}$ ) may be calculated (assuming a linear pH increase with depth) per Eq. 6.

$$C_{g,k} = 10^{\log_{10}(C_{g,k}^{\text{mean}}) - 1.33} \quad (6)$$

## 2.4 Effect factors

The effect factor (EF; [PDF  $\times$  (mol  $L^{-1})^{-1}$ ]) describes the change in potentially disappeared fraction of fish species (PDF) per unit of change in  $H^+$  concentration in lake  $k$ . Based on Payet (2006), the effect factor was calculated per Eq. 7:

$$EF_k = \frac{dPDF_k}{dC_k} = -\frac{1}{SR_{Ck}} \times \frac{dSR_k}{dC_k} = -\frac{1}{SR_{Ck}} \times \left[ \exp\left(a \times (-\log_{10}(C_k))^2 + b \times -\log_{10}(C_k) + c\right) \times \left(\frac{-1}{C_k \times \ln(10)} \times [2 \times a \times -\log_{10}(C_k)] + [b]\right) \right] \quad (7)$$

Where  $SR_{Ck}$  [dimensionless] is lake  $k$  fish species richness at the background surface concentration  $C_k$ , and  $a$ ,  $b$  and  $c$  [dimensionless] are regression coefficients. The  $a$  and  $b$  coefficients have the ability to shift the curve to the left or right of the species richness optimum. Like coefficient  $c$ , they may also increase or decrease the amplitude of the curve at the optimum.

PDF was evaluated from the decrease of spatially differentiated fish species richness, which was evaluated from  $H^+$  surface concentration and species richness regressions from Amarasinghe and Welcomme (2002). Regressions are available in the Electronic supplement 3 for several regions: North America, tropical America, temperate Europe and Asia, Africa and tropical Asia, which represent 62 % of total number of lakes in the world. Regression coefficients for the remaining 38 % were approximated by other regional coefficients based on similar Köppen-Geiger climate conditions (Kottek et al. 2006).

It should be noted that EF's regressions showed poor correlations ( $0.01 < R^2 < 0.42$ ) when linking lake surface  $H^+$  concentration to fish species richness. This can be explained by the numerous parameters that influence species richness such as temperature, lake area, depth, altitude and pH (Amarasinghe and Welcomme 2002). As such, the study regressions represent tendencies rather than correlations.

## 2.5 Region-specific characterization factors

Since the specific source location of the input and output processes compiled in the LCI phase are seldom known (Finnveden et al. 2009), we derived emission-weighted CFs at three additional resolution levels: country, continent and world—the latter considered as the global default CF. These CFs were evaluated according to Eq. (8). The ArcMap 9.3 GIS software was used to determine country and continental areas.

$$CF_r = \frac{\sum_r CF_{g,r} \times Em_{g,r}}{\sum_r Em_{g,r}} \quad (8)$$

Where  $CF_r$  is the CF at the specified resolution (country, continental and world),  $CF_{g,r}$  the CF at the  $2^\circ \times 2.5^\circ$  grid resolution inside the specified resolution  $r$  and  $Em_{g,r}$  the  $2^\circ \times 2.5^\circ$  emission inside the specified resolution  $r$ .

## 2.6 Regression analysis

The influence of the FFs, RFs and EFs on the spatial variability of the CFs was evaluated for each  $2^\circ \times 2.5^\circ$  grid via regression analysis. On a log scale, the spatial variabilities in  $\sum FF$  and  $\sum RF \times RF$  were compared to their endpoint CFs per pollutant. The following statistics were considered for the regression analysis: the slope of the log-linear regression, the coefficient of determination ( $R^2$ ) and the residual sum of squares (RSS). Correlation between the different factors would be expressed by a slope close to one, high coefficient of determination and low residual sum of squares.

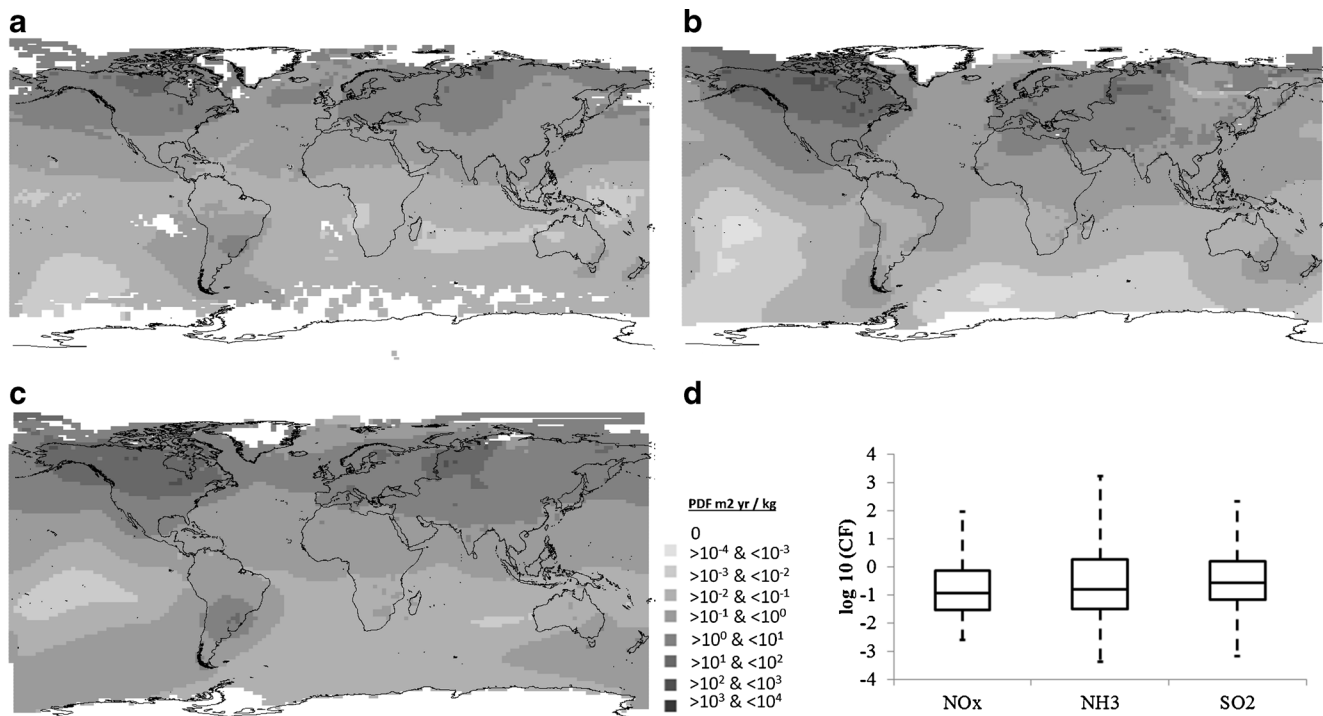
## 2.7 Parameter uncertainty analysis

The influence of FF, RF and EF on CF parameter uncertainty was evaluated by Monte Carlo simulation carried out in Matlab (v 7.10). The parameter uncertainty of each FF, RF and EF was calculated from lognormal distributions. Further details are provided in Electronic supplement 4. By randomly sampling (not Latin hypercube) 1,000 values for FF, RF and EF, we calculated 1,000 CFs for each of the 13,104 emitting grids (for a total of  $1,000 \times 13,104$  samples per pollutant), indicating the parameter uncertainty in the grid-specific CF values.

# 3 Results

## 3.1 Characterization factors

Figure 1 shows the spatially differentiated CFs (at  $2^\circ \times 2.5^\circ$ ) for the three acidifying pollutants considered. The highest CFs were found for emissions in Canada, Scandinavia and northern central Asia. The boxplot distribution shows that the median CF of  $SO_2$  is higher ( $0.26 \text{ PDF} \times \text{m}^2 \times \text{year} \times \text{kg}_{\text{emitted}}^{-1}$ ) than the median CF of  $NH_3$  ( $0.16 \text{ PDF} \times \text{m}^2 \times \text{year} \times \text{kg}_{\text{emitted}}^{-1}$ ) and  $NO_x$  ( $0.11 \text{ PDF} \times \text{m}^2 \times \text{year} \times \text{kg}_{\text{emitted}}^{-1}$ ). CF range over 5, 6 and 8 orders of magnitude for  $NO_x$ ,  $SO_2$  and  $NH_3$  emissions, respectively. As seen, the CF spatial variability is more important than the variability between pollutants. Grid-specific and global, continental and country averaged CFs are available in Electronic supplement 6, which shows that continental CF range over 3, 3 and 2 orders of



**Fig. 1** Worldwide spatially differentiated endpoint characterization factors for aquatic acidification at  $2^\circ \times 2.5^\circ$  grid resolution for emissions of **a**  $\text{NO}_x$ , **b**  $\text{NH}_3$  and **c**  $\text{SO}_2$ . **d** Box plot showing the distribution of the

calculated 13104 CFs. The *edge of the box* represents the 25th–75th percentiles, the *line in the box* represents the median and the *broken lines* represent the minimum and maximum of the distribution

magnitude and country CF range over 4, 6 and 5 orders of magnitude for emissions of  $\text{NO}_x$ ,  $\text{NH}_3$  and  $\text{SO}_2$ , respectively. Transcontinental deposition (provided in Electronic supplement 7) shows that, for most emission locations, the potential impacts related to aquatic acidification will occur on a different continent; only 30 to 45 % of the emission locations have 10 % or less of their potential impact occurring outside their respective continents.

### 3.2 Regression analysis

Table 1 presents the results of the regression analysis, which showed an explained variance of less than 0.21 for  $\sum \text{FF}$  towards CF. The explained variance of  $\sum \text{FF} \times \text{RF}$  towards CF results is an explained variance of 0.9 with linear regression slopes that approximate 1. The stronger correlation between  $\sum \text{FF} \times \text{RF}$  and the endpoint CFs is also confirmed by the relatively lower residual sum of squares of the regressions.

### 3.3 Parameter uncertainty

The results in Fig. 2 showed that, within a 95 % confidence interval, the ratios between Monte Carlo varied, and the original  $\sum \text{FF}$ ,  $\sum \text{FF} \times \text{RF}$  and CF were found, for all substances,

inside a  $10^{\pm 0.1}$  (0.79–1.26),  $10^{\pm 0.3}$  (0.50–2.0) and  $10^{\pm 2.0}$  (0.01–100) interval, respectively.

## 4 Discussion

### 4.1 Spatial variability

CFs vary by range over 5, 6 and 8 orders of magnitude for  $\text{NO}_x$ ,  $\text{SO}_2$  and  $\text{NH}_3$  emissions, respectively. This variation may primarily be explained by the locations of the lakes: the pollutant must deposit or travel to a lake to increase the value of a CF for a given emission location. Consequently, due to the extensive number of lakes in the northern latitudes, especially in Canada, Scandinavia and in the northern central Asia (i.e. the locations with the highest CFs), CFs for emission occurring in northern regions are typically higher than in the southern latitudes. This typically explains the crucial importance of RFs to spatial variations in the regression analysis and the difference in the range of CFs between pollutants. Indeed,  $\text{NO}_x$  has a wider deposition pattern (mostly continental) than  $\text{SO}_2$ , which in turn has a wider deposition pattern (mostly countries) than  $\text{NH}_3$  (mostly local). Thus, a more localized deposition near an extensive lake area will typically result in a



**Table 1** Regression analysis of the spatial variability between endpoint characterization factors (i.e.  $\sum FF \times RF \times EF$ ) with both atmospheric fate ( $\sum FF$ ) and atmospheric fate-receiving environment fate ( $\sum FF \times RF$ ) per emitted pollutants

	Equation of linear regression	Coefficient of correlation ( $R^2$ )	Residual sum of squares (RSS)
$\sum FF$ and CF (i.e. $\sum FF \times RF \times EF$ )			
NO <sub>x</sub>	$\log(CF) = 3.18 \log(\sum FF) + 26.00$	0.21	$5.91 \times 10^3$
NH <sub>3</sub>	$\log(CF) = -0.77 \log(\sum FF) - 6.87$	0.01	$1.25 \times 10^4$
SO <sub>2</sub>	$\log(CF) = 2.35 \log(\sum FF) + 18.71$	0.08	$8.14 \times 10^3$
$\sum FF \times RF$ and CF (i.e. $\sum FF \times RF \times EF$ )			
NO <sub>x</sub>	$\log(CF) = 0.81 \log(\sum FF \times RF) + 4.83$	0.90	$7.55 \times 10^2$
NH <sub>3</sub>	$\log(CF) = 0.87 \log(\sum FF \times RF) + 5.25$	0.89	$1.41 \times 10^3$
SO <sub>2</sub>	$\log(CF) = 0.81 \log(\sum FF \times RF) + 4.87$	0.90	$8.80 \times 10^2$

high CF unlike to the same localized deposition in an area with no lakes.

A high RF will occur when a small deposit results in a high increase in pH in a large lake, and a low RF will result from a high deposition with a low increase in pH in a small lake. Anything in between can be explained by the interaction of these parameters.

#### 4.2 Parameter uncertainty

The parameter uncertainty analysis showed that the addition of each new modelling step to calculate the respective intermediary parameters increased the uncertainty. However, EF is the major contributor to overall CF parameter uncertainty (98 % of the total uncertainty), while atmospheric and receiving environment fates each contributed to 1 %. The EF parameter uncertainty can be explained by the working point of the pH-species richness curve. Indeed, use of the lake mean H<sup>+</sup> concentration ( $C_{g,k}^{\text{mean}}$ ), which differs from the lake surface H<sup>+</sup> concentration ( $C_{g,k}$ ) by approximately 1.33 on a pH scale, may result in an increase of the CF by three or four orders of magnitudes.

#### 4.3 Limitations

Our uncertainty analysis only focused on parameter uncertainty. However, several other limitations or sources of uncertainties (outside the modelling algorithms) may affect the results of simulated CFs. This section addresses the most significant limitations.

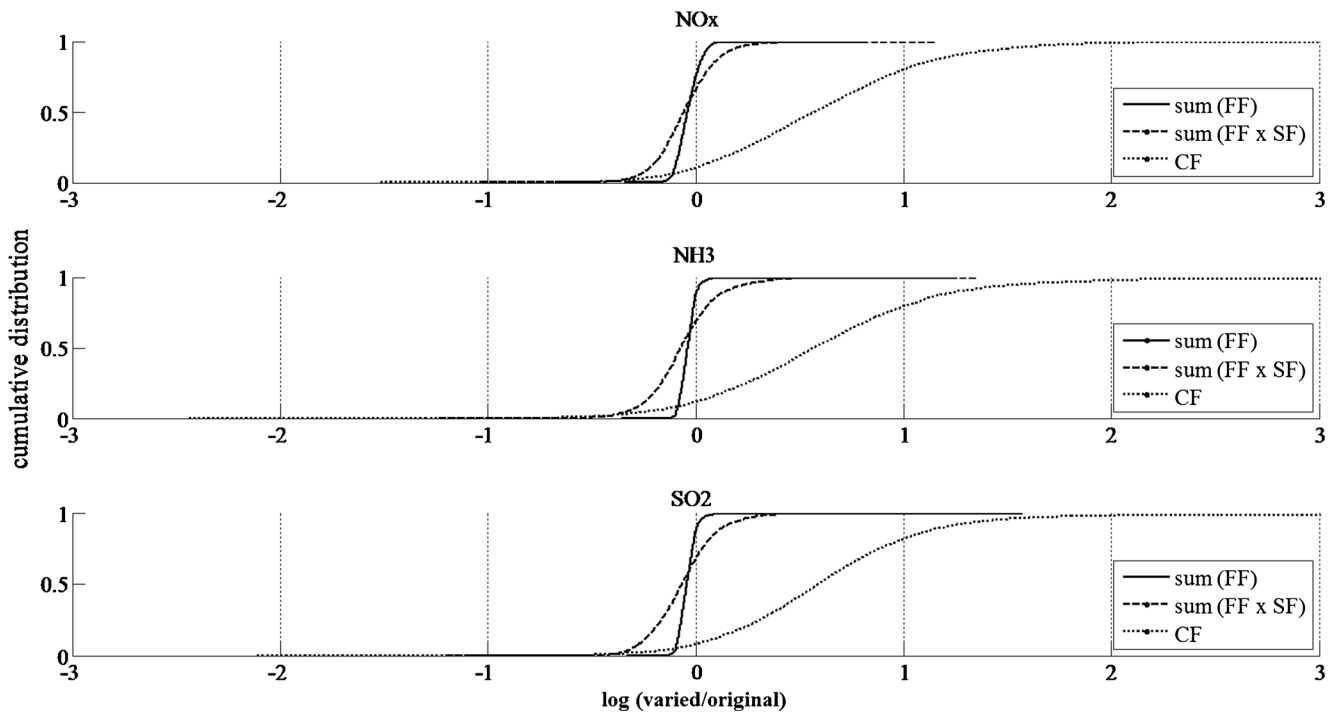
Firstly, it should be noted that the CFs were only designed to evaluate the potential acidification impacts in lakes. The aquatic acidification of rivers should not be evaluated with the CFs because of (1) the area of validity of Amarasinghe and Welcomme's regressions and (2) the adopted empirical corrections based on a default mean depth of 10 m are only valid for lakes. However, since worldwide river areas (610,760 km<sup>2</sup>) are significantly smaller than lake areas (2,427,630 km<sup>2</sup>) (Lehner and Döll 2004), we believe

that it is not paramount to include rivers since lakes are likely to be the major contributors to the RF and, consequently, the CFs.

Secondly, our model relies on a steady state approach and therefore neglects the acidification period (we cannot evaluate the timeframe in which a new equilibrium will be reached in a lake) and the seasonal or monthly variations of input parameters. The latter may be significant and would be more appropriate than yearly averaged parameters. For example, Driscoll and Van Dreaseon (1993) observed a seasonal variability of  $\pm 25$  % in a lake pH in the Adirondack. However, since LCA generally does not provide information on the time when emissions occur, the use of an annual parameter average is in line with the scope of the study. The acidification period can only be evaluated through dynamic modelling. However, considering the existing uncertainties and limitations, it would be ill-advised (for the time being) to add several new levels of difficulty inherent to dynamic modelling.

Thirdly, we neglected lake H<sup>+</sup> concentration variations within a  $0.5^\circ \times 0.5^\circ$  grid due to the lack of higher river network resolution in Vörösmarty et al. (2000a, 2000b). However, pH field measurements from 309 lakes over 150  $0.5^\circ \times 0.5^\circ$  grids in Canada showed that spatial variation within a grid was not uncommon: within a grid surface lake pH varied, on average, by 0.26 (on a pH scale) but could vary up to 1.2 (Electronic supplement 8). More precise data is required to improve the situation.

Fourthly, while terrestrial and air buffering capacities were considered, we neglected the biogeochemical reactions responsible for the production or consumption of H<sup>+</sup> ions in lakes (e.g. bedrock buffering capacities) due to global scale data limitations. We also believe that omitting these buffer reactions is insignificant over a long period of time in comparison to soil watershed inputs, as observed in eastern Canada (Jeffries and Ouimet 2004). To test the implications of this assumption, we randomly selected three different  $0.5^\circ \times 0.5^\circ$  grids and tested several values for  $r_{g,k}$  [volumes were evaluated based on data in Kourzeneva (2009)]. The tests showed an asymptotic behaviour close to a value of  $r_{g,k}$



**Fig. 2** Log 10 of the ratio between the Monte Carlo varied and original  $\sum FF$ ,  $\sum FF \times RF$  and CF (X-axis) for the cumulative distribution of  $2^\circ \times 2.5^\circ$  grids (Y-axis) for emissions of  $NO_x$ ,  $NH_3$  and  $SO_2$

specific to each lake (Electronic supplement 9). This is typically observed over a narrow range of  $r_{g,k}$  values specific to each lake. Otherwise, lake  $H^+$  concentration remained unchanged. Without a specific evaluation of  $r_{g,k}$  (whether or not it falls in the narrow section), the omission seems acceptable.

Fifthly, by using a CSTR assumption, we also neglected spatial variations within a lake. This represents a simplified estimation since lake pH levels are known to be highly heterogeneous. Indeed, a decrease of 2 to 3 pH scale units between the surface and bottom of the lake ( $\approx 10$  m) are common (Araoye 2009; Wang and Chen 1990). This heterogeneity prompted the use of an empirical correction to calculate surface pH from a lake mean pH. However, this correction is highly uncertain and significantly affects the CFs. The use of mean pH instead of surface pH increases CFs by three to four orders of magnitudes.

Considering the numerous model uncertainties, we wanted to ascertain the accuracy (or lack thereof) of lake pH simulations. To do so, we compared our simulated surface pH concentrations with lake surface field measurements from 309 lakes over 150  $0.5^\circ \times 0.5^\circ$  grids in Canada (Electronic supplement 8). It must be noted that field measurements are not necessarily at steady state but the simulated pH values are. Comparisons showed that 95 % of the simulated pH values were inside a  $\pm 25$  % interval of the estimated mean. These results echo those found by Roy et al. (2012a), who showed an

accuracy of  $\pm 25$  % in soil pH estimations. Considering that the same method was used in this paper and that soil inputs constitute major contributors, it was deemed acceptable to set out a  $\pm 25$  % variation in lake acidification in our model. Furthermore, a  $\pm 25$  % variation is in line with the  $\pm 25$  % seasonal variability of lake pH evaluated by Driscoll and Van Dreason (1993). We also evaluated the pH variations from our simulations in Europe ( $5.1 < pH < 11.2$ ; mean of 8.8), North America ( $5.2 < pH < 11.4$ ; mean of 8.2) and Africa ( $4.59 < pH < 11.6$ ; mean of 7.68). The simulated mean pHs of these regions were similar to the optimal pH based on the pH and species richness regressions in Amarasinghe and Welcomme: North America=8, Europe $\approx$ 8 (7.94 for temperate Europe and 8 for northern Europe) and Africa 8.2.

Sixthly, we focused our assessment on relative fish species richness, neglecting other types of freshwater species such as crustaceans or aquatic plants and insects. Some may argue that the *complex sensitivity of lakes* may not only be represented by fish. We lack the global level data required to approve or disprove this claim. However, based on the regional data collected in the province of Québec (Canada), fish species showed a tolerance to a change in lake acidity ( $4.5 < pH < 6.0$ ) similar to that of aquatic plants ( $4.5 < pH < 6.5$ ) and insects ( $4.0 < pH < 6.5$ ) (Dupont 2004).

Seventhly, we considered that there is no decrease in fish species if the pH is above the optimum (i.e. the pH at which species richness is highest). However, specific species may be

affected by changes in pH above the optimum. Therefore, our approach does not evaluate the effects of acidifying substances on specific species.

Finally, the quantification of a *marginal change* is still under discussion. Several authors (Huijbregts et al. 2000; Potting et al. 1998; Krewitt et al. 2001) used a +10 % variation, while others tried a combination of different values with a maximum of 50 % (Seppälä et al. 2006; van Zelm et al. 2007) when evaluating potential terrestrial acidification impacts. Similarly, Roy et al. (2012a) tested the consequences of acid deposit changes by increments of 1, 5 and 10 % with the same soil model used in this study to calculate  $C_{g,k}$ . They found a quasi-linear relationship between registered pH at 5 and 10 % increments. Without further counter indications, a 10 % increment was considered acceptable.

#### 4.4 Relationship to existing factors

We compared our European CFs to the CFs used in European life cycle impact assessment methods on the basis of potential impact contribution of the emission of 1 kg of each substance. The results of the comparison (Table 2) showed that the generic CFs have the same contribution per pollutant as our  $\Sigma FF$ . Discrepancies between the calculated  $\Sigma FF \times RF$  or CF and the generic CF can be explained by the latter's lack of receiving environment fate and/or effect factor.

We also found that our European CFs (1.4, 4.2 and 4.5  $PDF \times m^2 \times year \times (kg^{-1})$  for  $NO_x$ ,  $NH_3$  and  $SO_2$  emissions, respectively) are similar in terms of magnitude to the European endpoint terrestrial acidification in Van Zelm et al. (0.4, 1.5 and 0.5  $PNOF \times m^2 \times year \times (kg^{-1})$  for  $NO_x$ ,  $NH_3$  and  $SO_2$  emissions, respectively) and Goedkoop et al. (5.7, 15.6 and 1.0  $PDF \times m^2 \times year \times (kg^{-1})$  for  $NO_x$ ,  $NH_3$  and  $SO_2$  emissions, respectively). Further discrepancy discussions are moot since they represent different impact categories.

#### 4.5 Integration into LCA case studies

The characterization factors provided in our study enable the evaluation of the acidifying emissions impacts on spatially explicit relative fish species richness at the global scale. This

implies that the same emission occurring at two different locations can result in different impacts. Today, database providers such as ecoinvent (v3) (Weidema et al. 2012) are developing regional modules covering different regions of the world and addressing the geographic locations of emissions. In the event that the emission location is unknown or guessed, region-specific CFs on a country or continental scale or global default CFs may be used, also allowing for the evaluation of the uncertainty inherited from lack of spatial information.

## 5 Conclusions

This study provides the first spatially explicit CFs for aquatic acidification, overcoming the spatial limitations of current impact assessment methods and making it possible to consistently assess and discriminate emissions occurring in different geographic areas around the globe with the same characterization model. Nevertheless, this spatially explicit model introduces new limitations such as the inability to evaluate a river's acidification potential impacts or represent ecosystem sensitivity with species other than fish. Future research should focus on improving the effect evaluation by increasing the number of considered species and the accuracy of species richness regressions. Future research should also focus on decreasing the uncertainty modelling related to the fate in receiving environment pH evaluation.

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**Table 2** Comparison of the contribution to the total impact score when 1 kg of each substance are emitted

		Contribution[%]		
		Generic CFs from European methods	This study	
			$\Sigma FF$	$CF (\Sigma FF \times RF \times EF)$
$NO_x$	20		20	15
$NH_3$	52		52	31
$SO_2$	28		28	53



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